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(54) BENZOFURANONE AND INDOLINONE DERIVATIVES

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Abstract of the Disclosure JUL 19 1983 1150257

The present invention relates to a process for stabilising organic polymeric materials comprising incorporating therein a benzofuran(2)one compound or indolin(2)one compound containing at least two benzofuran-(2)one or indolin(2)one nuclei.

1150257

Case 150-4266/B

IMPROVEMENTS IN OR RELATING TO ORGANIC COMPOUNDS

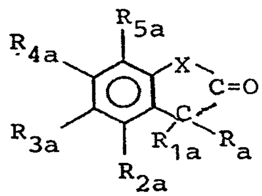
The present invention relates to a process for stabilizing organic polymeric materials employing benzofuranone or indolinone compounds as stabilisers.

Accordingly, the present invention provides a process for stabilising organic polymeric materials comprising incorporating therein a benzofuran(2)one or indolin(2)one compound contain at least two benzofuran(2)one or indolin(2)-one nuclei, respectively.

Preferred benzofuran(2)one compounds and indolin(2)-one compounds for use in the process of the present invention are bis-benzofuran(2)one or bis-indolin(2)one compounds in which the 3-position of the first benzofuran(2)one or indolin(2)one nucleus is bound directly to the 3- or 7-position of the second benzofuran(2)one or indolin(2)one nucleus, respectively, or the 5-, 6- or 7-position of the first benzofuran(2)one or indolin(2)one nucleus is bound directly to the same position of the second nucleus, and benzofuran(2)one or indolin(2)one compounds in which the 3-, 5-, 6-, or 7-position of the benzofuran(2)one or indolin(2)one nucleus is attached to the same position of 1 to 5 further such

nuclei through a 2 to 6 valent bridge member.

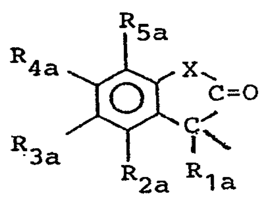
Preferred directly bound bis-benzofuran(2)ones and bis-indolin(2)ones are those of formula I_a,



I_a

in which X is -O- or $-\overset{|}{N}R_{10a}$

either, R_a is hydrogen or (aa /1)

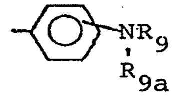


(aa/1)

and each R_{1a}, independently, is hydrogen; C_{1-22}-alkyl; C_{5-6}cycloalkyl; C_{1-5}alkyl-C_{5-6}cycloalkyl; phenyl; phenyl substituted by a total of up to three substituents selected from the group consisting of C_{1-12}alkyl (up to three of these with max. 18 carbon atoms in the combined alkyl substituents), hydroxyl (max. of two of these), C_{1-12}alkoxy, C_{1-18}acyloxy, chlorine and nitro (max. of one of each of these); a group of formula (a/4), (a/5) or (a/6)

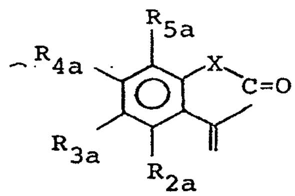
$$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-OR_7 \qquad (a/4)$$

$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-N(R_8)R_8$ (a/5)



(a/6)

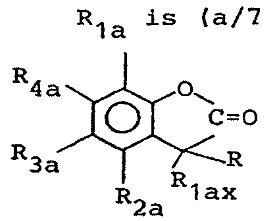
or, R_a together with R_{1a} is (a/3)



(a/3)

or, R_a is hydrogen and

R_{1a} is (a/7)



(a/7)

with the proviso that when R_{1a} is (a/7) X is -O-,

R_{1ax} is phenyl or phenyl substituted by a total of up to three substituents selected from the group consisting of C_{1-12}alkyl (max. three of these with a total of up to 18 carbon atoms in the combined alkyl substituents), hydroxyl (max. two of these), C_{1-12}alkoxy, C_{1-18}acyloxy, chlorine and nitro (max. one of each of these),

and R_{2a} to R_{5a}, independently, is hydrogen; C_{1-12}alkyl;
max. two of R_3 to R_5 are:
C_{5-6}cycloalkyl; C_{1-5}alkyl-C_{5-6}cycloalkyl; hydroxyl; C_{1-22}alkoxy; phenoxy optionally

substituted by up to two C_{1-12}alkyl groups with a total of up to 16 carbon atoms in the combined alkyl substituents; C_{1-18}-acyloxy; phenylcarbonyloxy; chlorine;

max. one of R_3 to R_5 is: -

phenyl-C_{1-9}alkyl or phenylthio in which the phenyl nucleus is optionally substituted by up to three substituents selected from C_{1-12}alkyl, hydroxyl, and $R_{15}CO-O-$;

phenyl optionally substituted by up to two C_{1-12}alkyl groups with a total of up to 16 carbon atoms in the combined substituents; nitro;

$-\overset{O}{\overset{\|}{C}}-R_{11}$ (b/2);

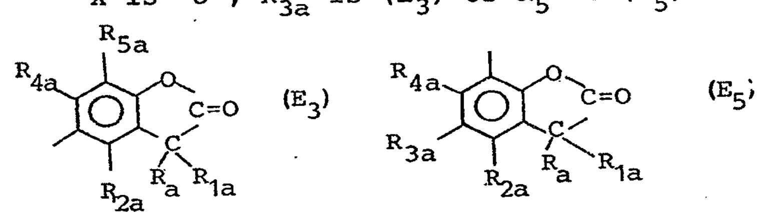
$-CH_2S-R_{12}$ (b/3) as R_3

$-CH(C_6H_5)CO-O-R_7$ (b/4) as R_3

(a/4) or (a/5) as R_3 or R_5

with the proviso that when R_{11} in (b/2) is other than hydrogen such (b/2) group is adjacent a hydroxyl group,

or, when R_a is hydrogen, R_{1a} is other than (a/7) and X is -O-, R_{3a} is (E_3) or R_5 is (E_5)



R_7, is C_{1-18}alkyl; alkyl-O-alkylene with a total no. of up to 18 carbon atoms; alkyl-S-alkylene with a total no. of up to 18 carbon atoms; di-C_{1-4}-alkylaminoC_{1-8}alkyl; C_{5-7}cycloalkyl; or phenyl optionally substituted by up to 3 C_{1-12}alkyl groups with a total no. of up to 18 carbon atoms in the combined substituents,

either, each R_8, independently, is hydrogen; C_{1-18}alkyl; C_{5-6}cycloalkyl; C_{1-5}alkyl$-C_{5-6}$cycloalkyl; phenyl optionally substituted by up to two C_{1-12}alkyl groups with max. 16 carbon atoms in the combined substituents;

$-CH_2CH_2OH$ (d/1) ;

$-CH_2CH_2OC_{1-18}$alkyl (d/2) ; or

$-CH_2CH_2-O-\overset{O}{\overset{\|}{C}}-R_{11}$ (d/3)

or, both R_8 together with the nitrogen form piperidine or morpholine,

R_9 has one of the significances of R_8,

R_{9a} is hydrogen, C_{1-18}alkyl, (d/1), (d/2) or (d/3),

R_{10a} is hydrogen, C_{1-18}alkyl, C_{5-6}cycloalkyl,

C_{1-5}alkyl-C_{5-6}cycloalkyl or phenyl optionally substituted by up to two C_{1-12}alkyl groups with max. 16 carbon atoms in the combined substituents, or benzyl,

R_{11} is hydrogen, C_{1-22}alkyl, C_{5-7}cycloalkyl, phenylC_{1-6}alkyl or phenyl optionally substituted by up to two C_{1-12}alkyl groups with max. 16 carbon atoms in the combined substituents,

R_{12} is C_{1-18}alkyl, 2-hydroxyethyl, phenyl or (C_{1-9})alkylphenyl,

R_{15} is C_{1-22}alkyl or phenyl, and

n is 0, 1 or 2,

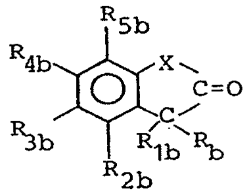
and the molecule contains only two benzofuran(2)one or indolin(2)one nuclei, whereby the substituents on the two benzofuran(2)one or indolin(2)one nuclei are the same or different, preferably they are the same.

Of the directly bound benzofuran(2)ones and indolin(2)-ones, the benzofuran(2)one compounds are preferred.

Preferred poly-benzofuran(2)ones or indolin(2)ones linked by a bridging group are those in which the bridging group is bound to the 3-, 5- or 7-position of the benzofuran(2)one or indolin(2)one nucleus.

When the bridging group is attached to the 3-position of the benzofuran(2)one or indolin(2)one nucleus, such group may be bound via a single or double bond.

Preferred bridged benzofuran(2)one or indolin(2)one compounds are those of formula I_b,



I_b

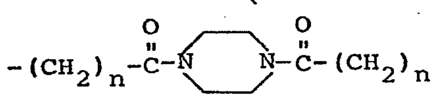
in which -X- is as defined above,

R_b, R_{1b}, R_{2b}, R_{3b}, R_{4b} and R_{5b} correspond to the significances R_a, R_{1a}, R_{2a}, R_{3a}, R_{4a} and R_{5a} above with the exception that the molecule is free from groups of formulae (aa/1), (a/3), (a/7), (E_3) and (E_5) <u>and</u>

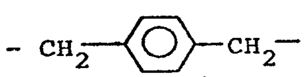
either R_{1b}, or R_b and R_{1b} together, or R_{3b}, or R_{5b} is bound to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei through a polyvalent bridge member.

Preferred groups in place of R_{1b} are: -

$$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-Z-A\left(-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_n\right)_w \qquad (e/1)$$

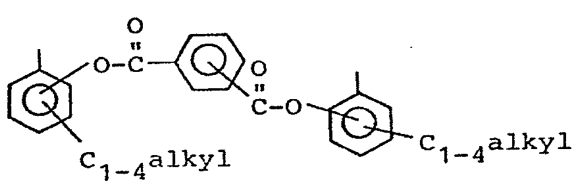


(e/4)

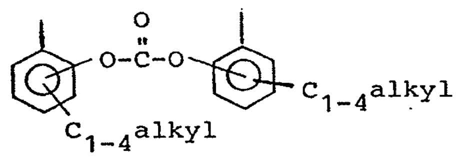


(e/5)

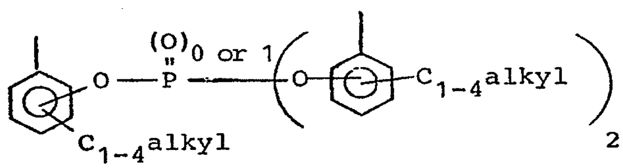
$$-C_mH_{2m}- \qquad (e/6)$$



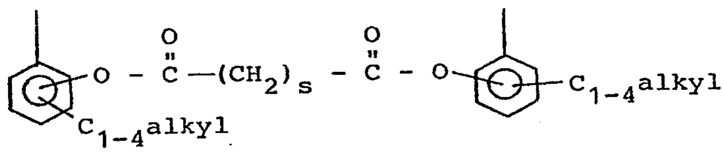
(e/7)



(e/7a)

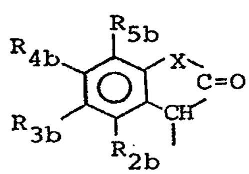


(e/7b)



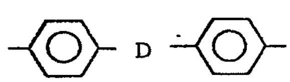
(e/8)

in which the free valencies are attached to groups E_1



E_1

A is a 2 to 6 valent saturated alkylene which optionally contains sulphur, oxygen, nitrogen or cyclohexylene bridges or is a 2- or 3-valent benzene radical or when both Z's are -O-, A is also (e/16)



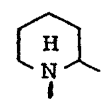
(e/16)

whereby when A is a 3-, 4-, 5- or 6-valent radical the further valencies are bound to OH, $-NHR_{10}$ or $-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_1$ groups, with the proviso that any free valencies on nitrogen in A itself are attached to $-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_1$,

w is from 1 to 6,

each Z, independently, is $-O-$ or $-\overset{|}{N}R_{10}$,

R_{10} has one of the significances of R_{10a} above or R_{10} together with the N-atom signifies

 or  ,

D is a direct bond or $-O-$, $-S-$, $-SO_2-$ $>C=O$ or $-\underset{R_{13}}{\overset{R_{13}}{C}}-$

in which each R_{13}, independently, is hydrogen, C_{1-16}alkyl (preferably C_{1-4}-alkyl) with the proviso that when both R_{13} are alkyl the combined groups contain max. 16 carbon atoms, phenyl, (a/4) or (a/5);

n is as defined above,

m is 2 to 10, and

s is 0 or 1 to 12.

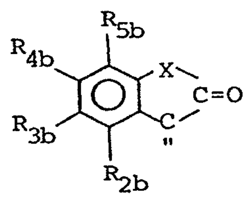
Preferred groups in place of R_b and R_{1b} together are: -

$$= \overset{R_{16}}{C} - \overset{O}{\overset{\|}{C}} - Z - A \left(Z - \overset{O}{\overset{\|}{C}} - \overset{R_{16}}{C} \right)_w \qquad (e/9)$$

$=HC \text{—} (C_pH_{2p}) \text{—} CH=$ (e/12)

 (e/13)

in which the free valencies are attached to groups E_{1a}

 E_{1a}

and A, Z, w and R_{10} are as defined above, with the exception that on A the further free valencies are attached to -OH, $-NHR_{10}$ or $-Z-\overset{O}{\overset{\|}{C}}R_{16}=E_{1a}$, and any free valencies on nitrogen, in A itself are attached to $-\overset{O}{\overset{\|}{C}}-CR_{16}=E_{1a}$,

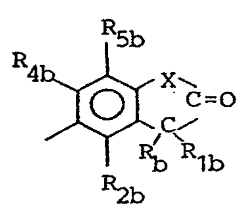
p is 0 or 1 to 10, and

R_{16} is hydrogen or methyl.

Preferred groups in place of R_{3b} are: -

$-O-$, $-S-$, $-SO_2-$, $>C=O$ or $-\underset{R_{13}}{\overset{R_{13}}{C}}-$ in which R_{13} is as defined above,

or (e/1), or (e/4) in which the free valencies are attached to groups E_{3b}

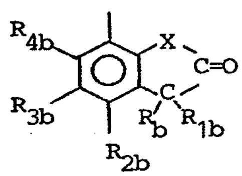
 E_{3b}

and A, w, Z and R_{10} are as defined above, with the exception that on A the further free valencies are attached to -OH, $-NHR_{10}$ or $-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_{3b}$ and any free valencies on nitrogen in A itself are attached to $-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_{3b}$.

Preferably when R_{3b} is a bridging group bound to one or more E_{3b} nuclei, X is -O- in all cases.

Preferred groups in place of R_{5b} are: -

-S- or $-\underset{R_{13}}{\overset{R_{13}}{C}}-$ in which R_{13} is as defined above, or (e/1), or (e/4) in which the free valencies are attached to groups E_{5b}



E_{5b}

and A, w, Z and R_{10} are as defined above, with the exception that the further free valencies on A are attached to -OH, $-NHR_{10}$ or $-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_nE_{5b}$ and any N-free valencies in A itself are attached to $-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_{5b}$.

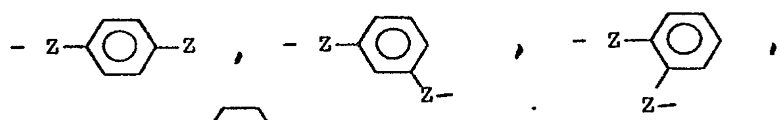
Preferably when R_{5b} is a bridging group bound to E_{5b}, X is -O- in all cases.

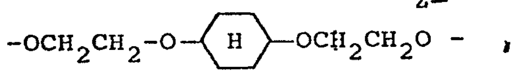
Examples of 2-valent -Z-A-Z- groups are: -

$-Z\ (CH_2)_{\overline{n_1}}\,Z-$ where n_1 is 2 to 12,

$-O(CH_2CH_2O)_{\overline{1,2\text{ or }3}}\,CH_2CH_2O-$

$-OCH_2CH_2SCH_2CH_2O-$, $-OCH_2CH_2S-SCH_2CH_2O-$,



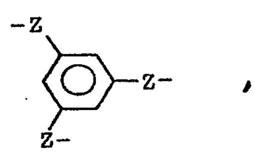


$-\overset{R_{10}}{\overset{|}{N}}-C_qH_{2q}O-$ where q is 2 or 3,

$-OCH_2CH_2\underset{R_{10}}{\underset{|}{N}}\ CH_2CH_2O-$,

$-O-CH_2-\overset{\text{alkyl}C_{1-4}}{\underset{\text{alkyl}C_{1-4}}{C}}-CH_2O-$, $Et\ \overset{NR_{10}}{C}-CH_2Z-$, $CH_3\overset{Z}{C}H-CH_2\overset{R_{10}}{N}-$

Examples of 3 valent -Z-A-Z- groups are: -

$C_{1-2}\text{alkyl}-\overset{CH_2-O-}{\underset{CH_2-O-}{C}}-NH-$, $-O-CH_2\underset{O}{CH}-CH_2-O-$, 

$-H\overset{|}{N}-(CH_2)_3-N(CH_2CH_2O-)_2$, $N(CH_2\overset{CH_3}{C}HO-)_3$, $-Z-CH_2\underset{C_{1-6}\text{alkyl}}{C}-(CH_2-Z-)_2$,

$N(CH_2CH_2O-)_3$

$-HN-(CH_2)_{\overline{2\text{ or }3}}\overset{|}{N}-CH_2CH_2-O-$, $HN-(CH_2CH_2-O-)_2$,

Examples of 4 valent -Z-A-Z- groups are: -

$(-OCH_2)_4-C$,

$-NHCH_2CH_2\overset{|}{N}CH_2CH_2\overset{|}{N}CH_2CH_2O-$, $\quad -\overset{H}{\overset{|}{N}}-\overset{CH_2O-}{\underset{CH_2-O-}{\overset{|}{\underset{|}{C}}}}-CH_2O-$,

An Example of a 5-valent -Z-A-Z- groups is: -

$-O-CH_2-(\overset{\overset{|}{O}}{\overset{|}{C}H})_3-CH_2-O-$

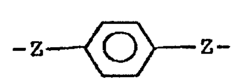
Examples of 6-valent -Z-A-Z- groups are: -

$-O-CH-(-\overset{\overset{|}{O}}{\overset{|}{C}H})_4-CH_2-O-$ and $[(-O-CH_2)_3-C-CH_2-]_2O$

A is preferably 2-, 3- or 4-valent with the following -Z-A-Z- groups being most preferred:

$-Z-(CH_2)_{n_1}-Z-$ with n_1 = 2 to 6 or 10

$-OCH_2CH_2OCH_2CH_2O-$,



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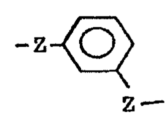
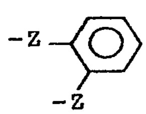
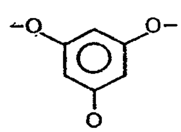




$-O-(CH_2CH_2O-)_{2 \text{ or } 3}CH_2CH_2O-$

$N(CH_2CH_2O-)_3$,



$-CH_2-\underset{C_{1-2}alkyl}{\underset{|}{C}}-(CH_2O)_2$ and $C-(CH_2-O-)_4$ $\quad -\overset{H}{\overset{|}{N}}-\overset{CH_2O-}{\underset{CH_2O-}{\overset{|}{\underset{|}{C}}}}-CH_2O-$

especially those in which A is alkylene.

Most preferred Z-A-Z groups are: -

$C(CH_2-O-)_4$ and $-O-(CH_2)_{n_1}O-$ with n_1 = 2 to 6 or 10

s is preferably 0 to 10 more preferably, 0 to 8.

m is preferably 2 to 6.

p is preferably 2 or 3, more preferably 3.

When R_{1b} is a bridge member carrying one or more benzofuran(2)one or indolin(2)one nuclei it is preferably R_{1b}' where R_{1b}' is (e/1), (e/4), (e/6), (e/7), (e/7a), (e/7b) or (e/8) more preferably (e/7) or (e/8).

When R_{1b} and R_b together form a bridge member carrying one or more benzofuran(2)one or indolin(2)one nuclei they preferably form (e/9) or (e/13),

When R_{3b} is a bridge member carrying one or more benzofuran(2)one groups it is preferably R_{3b}', where R_{3b}' is -S- or $-\overset{R_{13}'}{\underset{R_{13}'}{\overset{|}{\underset{|}{C}}}}-$, (e/1) or (e/4), where each R_{13}', independently, is hydrogen, C_{1-4}alkyl or (a/4) in which R_7 is hydrogen, C_{1-18}alkyl (preferably methyl) with the proviso that when one R_{13}' is (a/4) the other R_{13}' is other than (a/4) preferably methyl. More preferably R_{3b} as a bridging member carrying one or more benzofuran(2)one nuclei, is R_{3b}'' where R_{3b}'' is

$$-S-,\ -\overset{CH_3}{\underset{CH_3}{\overset{|}{\underset{|}{C}}}}-\ ,\ -CH_2-\ ,\ -CH_2-\ \text{(e/1) or (e/4), especially (e/1).}$$

When R_{5b} is a bridge member carrying one or more benzofuran(2)one groups it is preferably R_{5b}' where R_{5b}' is

$$-S-,\ -\overset{R_{13}'}{\underset{R_{13}}{\overset{|}{\underset{|}{C}}}}-\ \text{ where } R_{13}' \text{ is as defined above or (e/1).}$$

More preferably R_{5b} as a bridge member is R_{5b}'', where R_{5b}'' is -S- or $-CH_2-$.

In the compounds of formula I_b the substituents on each benzofuran(2)one or indolin(2)one nucleus are the same or different, preferably they are the same.

Of the benzofuran(2)one or indolin(2)one compounds having a bridge member bound to further such nuclei, the benzofuran(2)one compounds are preferred.

R_a is preferably R_a', where R_a' is hydrogen or (aa/1) with (aa/1) being most preferred. In (aa/1) preferably R_{1a} to R_{5a} have the preferred significances as stated herein.

In (a/7), R_{1ax}, R_{2a} , R_{2a}, R_{3a} and R_{4a} preferably have the preferred significances stated herein.

In (a/3) preferably R_{2a} to R_{5a} have the preferred significances stated herein.

R_{1a} is preferably R_{1a}', where R_{1a}' is (a/7) or R_1', where R_1' is hydrogen, C_{1-18}alkyl, phenyl optionally substituted by one or two C_{1-8}alkyl groups and/or a hydroxyl group; (a/4) or (a/5). More preferably R_{1a} is R_1'', where R_1'' is C_{1-18}alkyl or phenyl optionally substituted by one or two (C_{1-8})alkyl groups and/or a hydroxyl group. Most preferably R_{1a} is R_1''' , where R_1''' is phenyl optionally substituted by C_{1-4}alkyl, with unsubstituted phenyl being most preferred.

R_{1ax} is preferably R_1'', most preferably phenyl optionally substituted by C_{1-4}alkyl, with unsubstituted phenyl being most preferred.

R_{2a} and R_{2b} are preferably R_2', where R_2' is hydrogen

or C_{1-4}alkyl, more preferably R_2'' , where R_2'' is hydrogen or methyl, with hydrogen being especially preferred.

In E_3 preferably R_a, R_{1a}, R_{2a}, R_{4a} and R_{5a} have the preferred significances stated herewith.

R_{3a} is preferably R_{3a}', where R_{3a}' is E_3 or R_3', where R_3' is hydrogen or C_{1-9} (preferably C_{1-5})alkyl. Most preferably R_{3a} is R_3'.

R_{4a} and R_{4b} are preferably R_4', where R_4' is hydrogen or C_{1-4}alkyl, with hydrogen being especially preferred.

In E_5 preferably R_a and R_{1a} to R_{4a} have the preferred significances stated herein.

R_{5a} is preferably R_{5a}', where R_{5a}' is E_5 or R_5', where R_5' is hydrogen or C_{1-8}(preferably C_{1-5})alkyl. More preferably R_{5a} is R_5'.

R_b is preferably R_b' where R_b' is hydrogen or together with R_{1b} is (e/9) or (e/13).

R_{1b} is preferably R_{1bx}, where R_{1bx} is either R_1', more preferably R_1'' especially phenyl, or R_{1b}', especially with the preferred A groups in (e/1) and with s as 1 to 8, preferably 4 in (e/8).

R_{3b} is preferably R_{3bx}, where R_{3bx} is hydrogen, C_{1-9}alkyl or R_{3b}', more preferably hydrogen ,methyl or R_{3b}''

R_{5b} is preferably R_{5bx}, where R_{5bx} is hydrogen, C_{1-8}alkyl or R_{5b}', more preferably hydrogen or R_{5b}''.

X is preferably -O- or $-\dot{N}R_{10a}'$, where R_{10a}' is hydrogen, (C_{1-12})alkyl, phenyl or benzyl,more preferably, hydrogen, (C_{1-4})alkyl or phenyl. Most preferably X is -O-.

R_{10} is preferably R_{10}', where R_{10}' is hydrogen,

(C_{1-12})alkyl or phenyl, more preferably hydrogen or (C_{1-4})-alkyl, especially hydrogen or methyl.

R_7 in (a/4) and (b/4) is preferably R_7' where R_7' is hydrogen, C_{1-18}alkyl, phenyl optionally substituted by up to two C_{2-12}alkyl groups with max. 16 carbon atoms in the combined substituents. More preferably R_7 is R_7'', where R_7'' is C_{1-18}alkyl, phenyl or C_{1-12}alkylphenyl. Most preferably R_7 is C_{1-18}alkyl, especially C_{8-18}alkyl.

Each R_8, independently, is preferably R_8', where R_8' is hydrogen, C_{1-18}alkyl or both R_8's together form morpholine or piperidine. More preferably each R_8, independently is hydrogen or C_{1-18}alkyl. Preferred alkyl groups as R_8 are C_{1-12}-, preferably C_{1-8}-, most preferably C_{1-4}alkyl.

R_9 is preferably R_9', where R_9' is hydrogen, C_{1-8}-alkyl or (d/1). More preferably R_9 is hydrogen or C_{1-8}-alkyl. The preferred alkyl as R_9 contains 1 to 4 carbon atoms.

R_{9a} is preferably R_{9a}', where R_{9a}' is hydrogen or C_{1-8}-alkyl. Any alkyl as R_{9a} preferably contains 1 to 1 to 4 carbon atoms.

R_{11} is preferably R_{11}', where R_{11}' is hydrogen, C_{1-18} alkyl or phenyl. R_{11} in (b/2) is preferably phenyl.

Any alkyl as R_{11} preferably contain 1 to 17 carbon atoms.

R_{12} is preferably R_{12}', where R_{12}' is C_{1-12}alkyl, phenyl or 4-(alkyl C_{1-9})phenyl.

R_{16} is preferably hydrogen.

n in (a/4) or (a/5) as R_1 or R_5 is preferably 1.

n in (a/4) or (a/5) as R_3' is preferably 2.

Preferred compounds of formula I_a, are those in which X is -O- R_a is R'_a, R_{1a} is R'_{1a}, R_{2a} is R'_2, preferably R''_2, R_{3a} is R'_{3a}, R_{4a} is R'_4 and R_{5a} is R'_{5a} with the proviso that the molecule contains two and only two directly bound benzofuran(2)one nuclei.

More preferred compounds of formula I_a are those in which R_a is (aa/1), both R_{1a}'s are R''_1, more preferably R'''_1, especially phenyl, R_{2a} is hydrogen, R_{3a} is R'_3, preferably hydrogen, R_{4a} is hydrogen, R_{5a} is R'_5, preferably hydrogen and X is -O- and each benzofuran(2)one nucleus is identical.

When R_{3b} is a bridge member carrying further benzofuran(2)one nuclei preferably R_2 and R_4 are both hydrogen and R_{5b} is hydrogen or C_{1-4}alkyl, especially hydrogen.

When R_{5b} is a bridge member carrying further benzofuran(2)one nuclei preferably R_{3b} is hydrogen or C_{1-8}alkyl.

Preferred compounds of formula I_b are those in which R_b is R'_b, R_{1b} is R_{1bx}, R_{2b} is R'_2, preferably R''_2, R_{3b} is R_{3bx}, R_{4b} is R'_4, R_{5b} is R_{5bx} and X is -O-, and preferably each benzofuran(2)one nucleus is identical, with the proviso that only one bridge member bearing one or more benzofuran(2)one nuclei is present in the molecule.

More preferred compounds of formula I_b are those in which R_b is R'_b, R_{1b} is R_{1bx}, R_{2b} is hydrogen, R_{3b} is hydrogen, methyl or R''_{3b}, R_{4b} is hydrogen, R_{5b} is hydrogen or R''_{5b}, X is -O- and each benzofuran(2)one nucleus is identical.

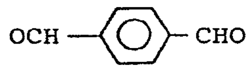
The directly bound bis benzofuran(2)one or indolin-(2)one compounds are either known or may be prepared in accordance with known methods from available starting materials.

The bridged benzofuran(2)one and indolin(2)one compounds as defined above are new and also form part of the present invention.

The bridged benzofuran(2)ones and indolin(2)ones of the invention may be prepared by conventional methods. For example, by condensing a polyfunctional bridge member with appropriately substituted benzofuran(2)ones or indolin(2)ones or by condensation and ring closure reactions of for example, hydroxy substituted bridged benzenes, and by interconversion reactions.

For example compounds of formula I_b in which one of R_1, R_3 or R_5 is (e/1) or (e/4), may be prepared by reacting a corresponding monomeric benzofuran(2)one compound or indolin(2)one compound in which one of R_1, R_3 and R_5 is $(CH_2)_n\overset{\overset{O}{\parallel}}{C}OH$ or a functional derivative thereof with a compound H-Z-A-$(Z\text{-}H)_w$ compound or with a di-or tri-aminobenzene or with a di- or tri-hydroxybenzene or for (e/4) with piperazine in known manner. Preferred functional derivatives are acid chlorides and lower alkylesters.

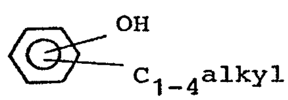
Compounds of formula I_b in which R_1 is (e/5) or (e/6) may be prepared by reacting a corresponding monomeric benzo-furan(2)one or indolin(2)one compound in which R and R_1 are both hydrogen with a compound of formula

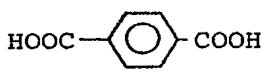
 or $OHC-C_mH_{2m}-CHO$

following by catalytic hydration in accordance with known methods.

Similarly, the compounds of formula I_b in which R and R_1 together form (e/9), (e/12) or (e/13) may be prepared by reacting the corresponding monomeric benzofuran(2)one or indolin(2)one compound where R and R_1 are both hydrogen with the corresponding aldehyde of each of the bridge members.

The compounds of formula I_b in which R_1 is (e/7), (e/7a) (e/7b) or (e/8) can be prepared by reacting the corresponding monomeric benzofuran(2)one or indolin(2)one compound in which R_1 is



with  for (e/7)

or a functional derivative thereof,

with $HOOC-(CH_2)_s-COOH$ for (e/8)

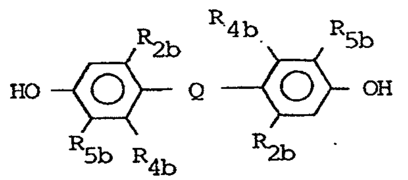
or a functional derivative thereof,

with $COCl_2$ for (e/7a), and

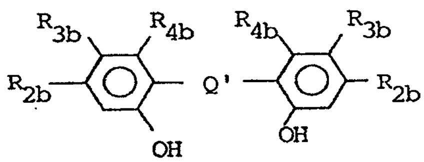
with $[O]_{0,1}\ PCl_3$ for (e/7b),

in accordance with known methods. Preferred functional derivatives are acid chloridesand lower alkyl esters.

The compounds of formula I_b, in which R_{3b} is $-O-E_{3b}$, $-S-E_{3b}$, $-SO_2E_{3b}$, $-\overset{O}{\overset{\|}{C}}-E_{3b}$, $-\overset{(R_{13})_2}{C}-E_{5b}$, or R_{5b} is $-S-E_{3b}$ or $-\overset{(R_{13})_2}{C}-E_{5b}$ by reacting a compound of formula III or IV



III for R_{3b}



IV for R_{5b}

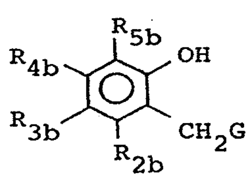
with a compound of formula $R_{1a}-\overset{OH}{\dot{C}H}-C-OH$ in a 1:2 molar ratio,

where Q is $-O-$, $-S_1-$, $-SO_2-$, $C{=}O$ or $-\overset{(R_{13})_2}{\dot{C}}-$

Q_1 is $-S-$ or $-\overset{(R_{13})_2}{\dot{C}}-$, and

R_1 is optionally substituted phenyl as given for R_{1b} above, by known methods.

The compounds of formula I_b in which R and R_1 are both hydrogen may be prepared by reacting a compound of formula V



V

in which R_{2b} to R_{5b} are as defined above and one of R_{3b} or R_5 is a bridge member as defined above linked to a further such phenolic compound, and with the exception that in any group (a/4) or (b/4), R_7 is hydrogen, and R_{3b} and R_{5b} are other than (a/5),

G is a secondary amine group or halogen,

with an ionic cyanide compound,

hydrolysing the product thereof,

followed by a ring closure condensation.

G is preferably $-N(C_{1-4})$alkyl or morpholine, especially $-N(CH_3)_2$. Any halogen as G is preferably chlorine or bromine, especially chlorine. Suitable ionic cyanide compounds are alkali- or alkaline earth cyanides, preferably sodium- or potassium cyanide. Each of the reaction steps may be carried out in accordance with known methods for such reactions.

The end product may be esterified or etherified to obtain compounds where R_7 is other than hydrogen. Furthermore, the methylene group in the 3-position may be reacted further to obtain compounds where R_1 is other than hydrogen.

The compounds of formula III, IV and V are either known or may be prepared by known methods from available starting materials. The same applies for the other starting materials.

The directly bound bis-benzofuran(2)ones, bis-indolin(2)one and bridged benzofuran(2)ones or indolin(2)ones as defined above (hereinafter referred to as compounds K) may be incorporated into the polymeric material to be stabilized before, during, or after polymerization.

The amount of compound K incorporated may vary according to the material to be stabilized and the ultimate use to which it is to be put. Suitable amounts are from 0.01 to 5% preferably from 0.05 to 1%, based on the weight of the materials to be stabilized. The organic polymeric materials to be stabilized may be natural or synthetic polymeric materials. Examples of such materials include rubber, polyolefins, especially polyethylene, polypropylene, ethylene/propylene copolymers, polybutylene, polystyrene, chlorinated polyethylene, PVC, polyester, polycarbonate, polymethylmethacrylate, polyphenyleneoxide, polyamides such

as nylon, polyurethanes, polypropyleneoxide, phenol-formaldehyde resins, epoxy resins, polyacrylonitrile and corresponding copolymers such as acrylonitrile butadiene styrene (ABS) terpolymers.

The process of the present invention is preferably employed to stabilise polypropylene, polyethylene, ethylene/propylene copolymers,PVC, polyesters, polyamides, polyurethanes, polyacrylonitrile, ABS terpolymers, terpolymers of acrylic ester, styrene and acrylonitrile, copolymers of styrene and acrylonitrile, styrene/butadiene copolymers, polybutylene and polystyrene. The most preferred organic polymeric materials are polypropylene, polyethylene especially HDPE, ethylene/propylene copolymers and ABS.

The incorporation of the compounds K in the material to be stabilized is effected in accordance with known methods. Preferred methods are those in which the compounds K are incorporated in the polymeric material by melt blending the stabiliser and the additives in conventional equipments such as Banbury mixers, extruders etc. Polypropylene and polyethylene granulates on powders are advantageously employed, whereby the compounds of formula I are admixed with said powders and then extruded etc and worked into the films, foils, bands threads etc.

The process of the present invention may be carried out by incorporating a compound K alone or together with other additives e.g. further stabilisers etc.

The preferred process according to the present invention comprises incorporating a compound K and either (i) a stabiliser of the sterically hindered phenol type,

or (ii) a sulphur-containing or phosphorus containing stabiliser,

or (i) and (ii), into the polymeric material.

The ratio of stabiliser (i) or (ii) to the compounds K incorporated in the polymeric material is suitably 5:1 to 1:5, preferably 2:1 to 1:1. The ratio of combined (other) stabilisers to compounds of formula I is suitably 15:1 to 1:5, preferably 6:1 to 1:3. Preferably, when only stabilisers (i) are employed with the compounds of formula I_c the ratio of compounds (i) to those of formula I_c is 3:1 to 1:1.

Examples of sterically hindered phenols are: - β-(4-hydroxy-3,5-ditert.-butylphenyl)-propionicacidstearyl-ester, tetrakis[methylene-3(3',5'-ditert.-butyl-4-hydroxy-phenyl)-propionate]-methane, 1,3,3-tris-(2-methyl-4-hydroxy-5-tert.-butylphenyl)-butane, 1,3,5-tris(4-tert.-butyl-3-hydroxy-2,6-dimethylbenzyl)-1,3,5-triazine-2,4,6-(1H,3H,5H)-trione, bis-(4-tert.-butyl-3-hydroxy-2,6-dimethylbenzyl)-dithiolterephthalate, tris(3,5-ditert.-butyl-4-hydroxybenzylisocyanurate, triester of 3,5-di-tert.-butyl-4-hydroxyhydrocinnamic acid with 1,3,5-tris-(2-hydroxyethyl)-s-triazin-2,4,6-(1H,3H,5H)-trione,bis [3,3-bis-4'-hydroxy-3-tert.-butylphenyl)-butaneacid]-glycolester, 1,3,5-trimethyl-2,4,6-tris-(3,5-ditert.-butyl-4-hydroxybenzyl)-benzene, 2,2'-methylene bis (4-methyl-6-tert.-butylphenyl) terephthalate, 4,4-methylene-bis-(2,6-ditert.-butylphenol), 4,4'-butylidene-bis-(6-tert.-butyl-meta-cresol), 4,4-thio-bis(2-tert.-butyl-5-methyl-phenol), 2,2'-methylene-bis(4-methyl-6-tert.-butylphenol

Examples of sulphur containing stabilisers are distearyl-

B

thiodipropionate, dilaurylthiodipropionate, tetrakis(methylene-3-hexylthiopropionate)-methane, tetrakis (methylene-3-dodecyl-thiopropionate)-methane and dioctadecyldisulphide.

Examples of phosphorus containing compounds are trinonyl-phenylphosphite, 4,9-distearyl-3,5,8,10-tetraoxadiphosphaspiroun-decane, tris-(2,4-ditert.-butylphenyl)phosphite and tetrakis(2,4-ditert.butylphenyl)-4,4'-diphenylene diphosphonite.

In addition to the above further stabilisers, U.V. absorbers as described in DOS 2 606 358 e.g. 2-(2'-hydroxyphenyl)-benztri-azole, 2-hydroxybenzophenone, 1,3-bis(2-hydroxybenzoyl)benzene, salicylates, cinnamic acid esters, hydroxybenzoic acid esters, sterically hindered amines and oxylic acid diamides. Suitable such compounds are described in DOS 2 606 358.

Metal deactivators for example N,N'-dibenzoylhydrazide, N-benzoyl-N'-salicyloylhydrazide, N,N'-distearylhydrazide, N,N'-bis-[3-(3,5-ditert.-butyl-4-hydroxyphenyl)-propionyl]-hydrazide, N,N'-bis-salicyloylhydrazide, oxalylbis-(benzyl-idenehydrazide), N,N'-bis(3-methoxy-2-naphthoyl-)hydrazide, N,N'-di-α-phenoxy-butyloxy (isophthalyl-dihydrazide) may also be incorporated into the polymeric material.

Additional conventional additives may also be employed for example, flame retardants, antistatic agents etc.

Furthermore, an optical brightener may be incorporated in the polymer to be stabilised and so that the distribution of the additives which are intimately admixed with said optical brightener may be ascertained by fluorescence intensity measurements.

The present invention also provides master batches

B

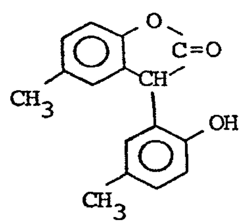
of polymeric organic materials containing 5 to 90%, preferably 20 to 60%, more preferably 20-30% of a compound K. Such master batches may then be admixed with unstabilised polymeric material. It is to be appreciated that such master batches may also contain additional additives such as those stated above.

Polymeric materials containing a compound K are primarily stabilised against degradation during processing. When, of course, other additives such as antioxidants, e.g. above phenols, and U.V. absorbers are also employed together with the compounds K the polymeric material has an enhanced long term stability against thermal- and photoxidative degradation.

The following examples further serve to illustrate the invention. In the examples all parts are by weight, and all temperatures are in degrees Centigrade.

Example 1

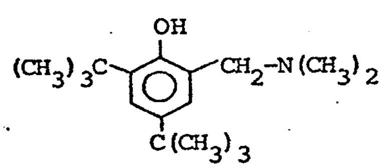
2.54 parts of the compound of formula



(prepared in accordance with known methods) and 1 part mandelic acid are heated to 200°C for 16 hours. Afterwards the reaction mixture is separated by column chromatography (silicagel, ether/petroliumether 1:2). Crystals having a melting point range of 185-187°C, corresponding to the formula of Compound No. 5 of the Table are obtained.

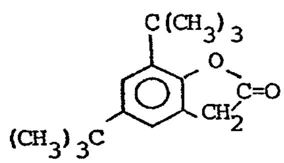
Example 2

78.9 Parts of the compound of formula



are dissolved in 450 parts diethyleneglycolmonomethylether. 39 Parts potassium cyanide and 6 parts potassium iodide are added thereto. At a temperature of 80°C, 63 parts of water are added dropwise. The temperature is raised to 130° and the mixture is stirred for 16 hours at this temperature. After cooling to room temperature, 1000 parts ice water are added. After carefully acidifying with

hydrochloric acid, a precipitate is formed which is dissolved in 400 parts ether. The organic phase is separated, washed with water, dehydrated over $MgSO_4$ and evaporated. The residue is added to toluene, heated to the boil for approximately 1 hour whereupon water of condensation separates out. After evaporating the solvent and recrystallizing from methanol a colourless crystalline product of formula



is obtained. A mixture of 19.36 parts of the compound, 5.36 parts terephthalic aldehyde, 0.24 parts piperidine benzoate and 100 parts toluene are heated for 15 hours at reflux temperature. After evaporation of the solvent the product is recrystallized from acetone. The so-obtained crystals are washed with a small amount of ice-cold ether and dried. A yellow powder having a melting point range of 241-242°C corresponding to the formula of Compound No. 9, is obtained.

Example 3

2,0 Parts pentaerythritol-tetra-[3-(4-hydroxyphenyl)-propionate] and 2,1 parts mandelic acid are heated together to 180°C for 23 hours. After cooling, the reaction mixture is separated by column chromatography (silicagel, eluent 9:1 toluene/acetone). The so-obtained product has a melting point range of 90-95°C, and

corresponds to the formula of Compound No. 10, Compound Nos. 1, 2 and 4 are made in analogous manner, starting with the corresponding bis phenol compound and reacting the same with mandelic acid.

Example 4

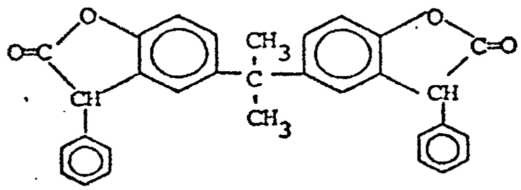
A solution of 2,0 parts terephthalic acid dichloride in 40 parts toluene is added slowly at room temperature to a mixture of 5,1 parts of the starting material used in Example 1, 100 parts toluene and 2,1 parts triethylamine. A white precipitate is obtained. The mixture is stirred for some hours at room temperature followed by stirring at 80°C for 2 hours. The precipitate is filtered off and the clear solution is evaporated. A white crystalline product, melting point 245-246°C, (recrystallized from acetone/petroliumether) of the formula of Compound No. 6 is obtained. Compound No. 7 can be prepared in analogous manner.

Example 5

1,60 Parts of Compound No. 9 are catalytically dehydrogenated at room temperature under normal pressure. As solvent 20 parts of glacial acetic acid is used, as catalyst 0.2 parts palladium on barium sulfate. After removal of the catalyst and the solvent, the residue is taken up with ether. The etherified solution is shaken with a Na-bicarbonate solution and then with water, followed by dehydrating over $MgSO_4$ and then evaporated. The product, having a melting point range of 258-259°C (petroliumether),

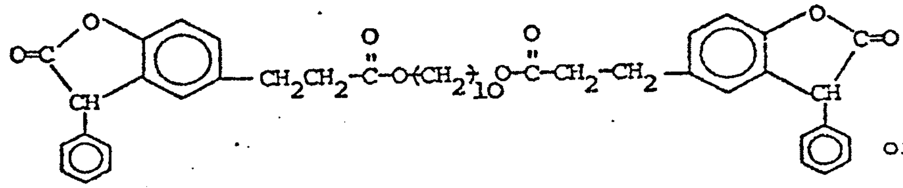
corresponds to the formula of Compound No. 8. Compound No. 3 of the Table is prepared in accordance with known methods.

No. 1



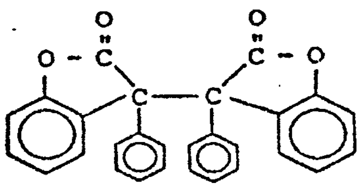
m.pt. 87-118°

No. 2



oil

No. 3



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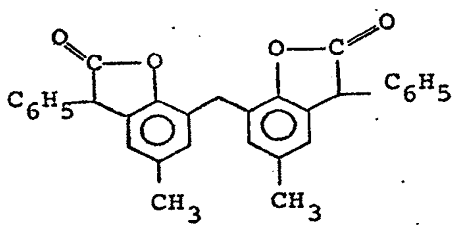
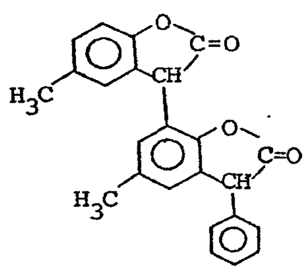
No. 4



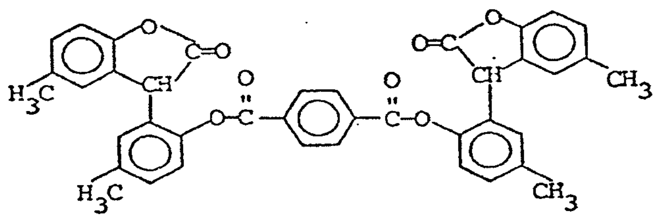
m.pt. 163.6 - 166°C

No. 5



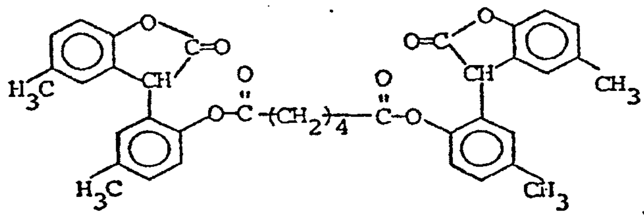
m.pt. 185-187°

No 6



m.pt. 244-5°

No. 7



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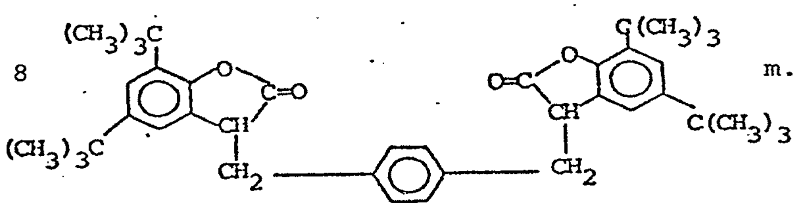
No. 8



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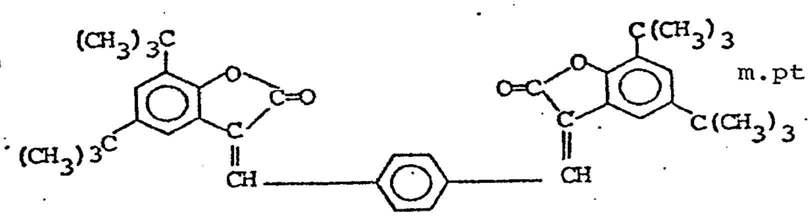
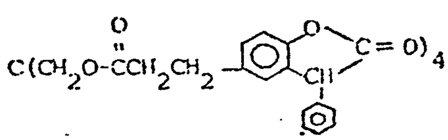
No. 9



m.pt. 241-242°

No. 10



m.pt. 90-95°

Example A

A mixture of 1200 parts of a commercially available unstabilized polypropylene (Profax® 6501), 0.6 parts calciumstearate, 0.6 parts tetrakis-[methylene-3(3',5'-di-tert.-butyl-4'-hydroxyphenyl)-propionate]-methane and 0.6 parts of the Compound No. 8 of the Table are shaken together for 10 minutes and extruded at 120 revs/min with temperatures of 150, 240, 260, and 200° in the different heating areas of the extruder to form a strand which is granulated after passing through a water bath. The granulate is extruded and granulated a further 9 times, each time a part is taken to measure the Melt Flow Index (MFI according to ASTM D 1238 L, 230°; 216 kg) which serves as a measure of the thermomechanical oxidative degradation of a polymer. A control without Compound 8 of the Table is also extruded in like manner and tested. In comparison, the polymer containing Compound No. 8 of the Table exhibits a greatly improved melt stability during continuous extrusion. The other compounds of the Table may be employed in like manner.

Example B

100 Parts unstabilized HD-polyethylene powder (Phillips Type) are stabilized with 0.02 parts Compound No. 2 of the Table and 0.01 parts tetrakis-[methylene-3(3',5'-di-tert.-butyl-4'-hydroxyphenyl)-propionate)-methane. The powder is subjected to a modified MFI Test at 230°/0.325 kg on a Davenport- MFI apparatus. The powder is pushed into a heated steel cylinder and a 325 g weight is placed

B

thereon. The polymer which is pressed out is cut off at 60 second intervals. The amount is calculated in terms of g/10 min. The stronger the crosslinking of the polymer owing to insufficient stabilization, the lower the MFI value. After 5 to 15 minutes a constant value is obtained. The other compounds of the Tables may be used in analogous manner.

Example C

1.0 Part octylstearate, 1.5 parts Ba-Cd stabilizer (powder-forming), 1 part of Compound No. 3 of the Table and 0.5 parts of a commercially available arylalkylphosphate are mixed with 100 parts commercially available dispersion PVC (k-value-60) in a Fluid Mixer (Papenmeier Type TEHK8) until the temperature has risen to 110°. The homogeneous mixture is rolled on rollers heated to 180° for 1 minute and then pressed into plates (thickness 1 mm) at 200° for 1.5 min. at 2 atm. and 1.5 minutes at 20 atm. The test or plates are put into an air circulating drying cabinet at 180°C for 30 minutes. A comparison sample which contained 2.5 parts Ba-Cd stabilizer instead of Compound No. 4 and 1.5 parts of the Ba-Cd stabilizer was also treated in the same manner. This sample undergoes discolouration even at the beginning of the heat treatment and is markedly more discoloured after the 30 minutes than the sample containing Compound No. 4 of Table 1.

Example D

300 Parts ABS powder (Fa. Marbon AOE 30/075) are

dissolved in 2200 parts chloroform and the solution is dropped into 8000 parts methanol whereupon the ABS is precipitated. After filtration the polymer which is now free from stabilizer is treated in vacuo overnight to remove all the solvent. 100 parts of the so-treated ABS powder is dissolved in chloroform and 0.2 parts Compound No.2 of the Table are added thereto and the whole is stirred under nitrogen atmosphere for 15 minutes. The solution is drawn into a film with a 1 mm doctor blade onto a glass plate and is left for the solvent to evaporate-off whereby the film shrinks to 150 μ thickness and is freed from the rest of the solvent overnight at room temperature in vacuo. The film is then stoved in an air-circulating oven at 95°. By repeated IR-measurement to $\Delta \varepsilon = 0.4$ at 1715 cm^{-1} the ageing resistance is checked. The samples containing the benzofuranone compound have longer resistance than the control samples which contain no stabilizer.

Example E

100 Parts granulated polyethyleneterephthalate are ground to a rough powder and dried overnight at 100° in a vacuum drying cabinet. 1.0 Part of Compound No. 2 of the Table is added and the mixture is homogenised, then granulated in an extruder, spun into fibres at 280°, stretched (120 den/14) and twisted. The fibres are wound on to white cards and exposed to the light in an Atlas Weatherometer for 24 hour intervals. In comparison to a non-stabilized control, the sample containing Compound No. 41 has less tendency to yellow during the exposure to light

and can be left in the Weatherometer for a substantially longer period of time in order to reach the same decrease in the tensile strength (50%).

Example F

1000 Parts 20% styrene-butadiene rubber emulsion are added with stirring to a hydrochloric acid 5% sodium chloride solution whereupon the rubber coagulates. Stirring is continued for 1 hour at pH 3,5. After filtration the coagulate is repeatedly washed and dried to a constant weight at room temperature in a vacuum cabinet.

25 Parts of this rubber are heated under nitrogen atmosphere to 125° in Brabender plastographs and mixed with 0.25 parts Compound No. 2 of Table 1 for 10 minutes and subsequently pressed to (0.5 mm thick) plates at 125°. The plates were put into an Atlas Weatherometer for 24 hour intervals together with samples containing no stabilizer. In comparison to the latter samples, the stabilized samples exhibited significantly better resistance to light.

Example G

49.5 Parts Compound No. 2 of the Table, 49.5 parts tetrakis-[methylene-3-(3',5'-di-tert.-butyl-4'-hydroxyphenyl)-propionate]-methane, 1 part calcium stearate and 0.02 parts (7-[24-naphthol(1,2d)triazol-2-yl]-3-phenylcumarine (optical brightener) are heated to 140°. The mixture melts with stirring and the melt is poured into a flat dish and ground after cooling. The product obtained melts at 70-75°C.

0.5 Parts of the ground melt are mixed in a plastic

bag by repeated shaking with 1000 parts unstabilized HDPE powder (Ziegler Type, MFI 190/z = 0.7). 43 Parts of the powder mixture are heated to 220° in a Brabender Plasti-Corder PLV 151 extruder at 50 revs/min.until there is a sharp drop in the torque indicating degradation (to cross-linking decreases). The test sample has good stability.

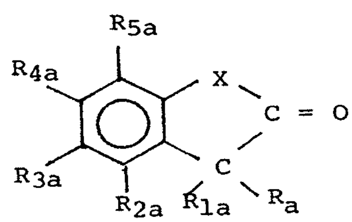
When different concentrations of the above melt product are mixed with polyethylene or polypropylene powder and extruded into a strand which are subsequently ground, the fluorescence intensity can be measured to assess the distribution of additives in the polymer mixture.

The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. A process for stabilising organic polymeric materials comprising incorporating therein a benzofuran(2) one compound or indolin(2)one compound containing at least two benzofuran(2)one or indolin(2)one nuclei.

2. A process according to Claim 1, in which either a bis-benzofuran(2)one or bis-indolin(2)one compound in which the 3-position of the first benzofuran(2)one or indolin(2)one nucleus is bound directly to the 3- or 7-position of the second benzofuran(2)one or indolin(2)one nucleus, respectively, or the 5-, 6- or 7-position of the first benzofuran(2)one or indolin(2)one nucleus is bound directly to the same position of the second nucleus or a benzofuran(2)one or indolin(2)one compound in which the 3-, 5-, 6- or 7-position of the benzofuran(2)one or indolin(2) one nucleus is attached to the same position of 1 to 5 further such nuclei through a 2 to 6 valent bridge member is incorporated in the polymeric material.

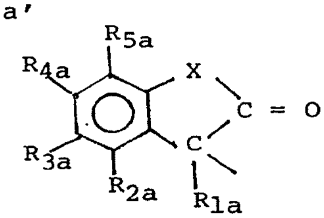
3. A process according to Claim 2 in which the directly bound bis-benzofuran(2)one or bis-indolin(2)one compound is of formula Ia,



Ia

in which X is -O- or $-\overset{|}{N}R_{10a}$,

either i) R_a is (aa/1)



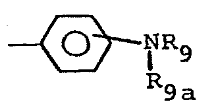
(aa/1)

B

and each R_{1a}, independently, is hydrogen; C_{1-22} alkyl; C_5 or C_6 cycloalkyl; C_{1-5} alkyl-C_5 or C_6 cycloalkyl; phenyl; phenyl substituted by one to three substituents selected from the group consisting of C_{1-12} alkyl, hydroxy, C_{1-12} alkoxy, C_{1-18} acyloxy, chloro or nitro, with the provisos that: 1) when the phenyl ring contains more than one C_{1-12} alkyl group, said alkyl groups contain a maximum of 18 carbon atoms, 2) the maximum number of hydroxy substituents is two, and 3) the maximum number of each of the substituents selected from C_{1-12} alkoxy, C_{1-18} acyloxy, chloro and nitro is one; or a group of formula (a/4), (a/5) or (a/6)

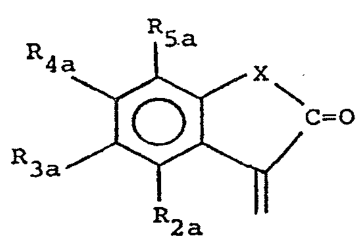
$$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-OR_7 \qquad (a/4)$$

$$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-\overset{R_8}{\overset{|}{N}}R_8 \qquad (a/5)$$



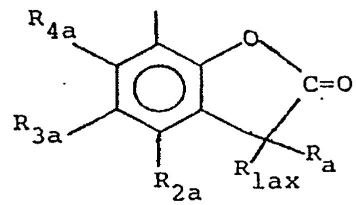
(a/6);

or ii) R_a together with R_{1a} is a group of formula (a/3)



(a/3);

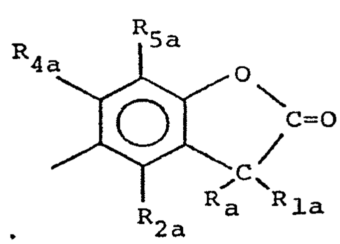
or iii) R_a is hydrogen, X is -O- and R_{1a} is a group of formula (a/7)



(a/7)

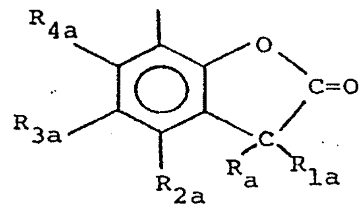
where R_{1ax} is phenyl; or phenyl substituted by one to three substituents selected from the group consisting of C_{1-12} alkyl, hydroxy, C_{1-12} alkoxy, C_{1-18} acyloxy, chloro or nitro, with the provisos that: 1) when the phenyl ring contains more than one C_{1-12} alkyl group, said alkyl groups contain a maximum of 18 carbon atoms, 2) the maximum number of hydroxy substituents is two, and 3) the maximum number of each of the substituents selected from C_{1-12} alkoxy, C_{1-18} acyloxy, chloro and nitro is one;

or iv) R_a is hydrogen, X is -O-, R_{1a} is other than a group of formula (a/7) and either R_{3a} is a group of formula (E_3)



(E_3)

or R_{5a} is a group of formula (E_5)



(E_5);

each of R_{2a} to R_{5a}, independently, is hydrogen; C_{1-12} alkyl; C_5 or C_6 cycloalkyl; C_{1-5} alkyl-C_5 or C_6 cycloalkyl; hydroxy; C_{1-22} alkoxy; phenoxy; phenoxy substituted by one or two C_{1-12} alkyl groups, said alkyl groups having a maximum of 16 carbon atoms; C_{1-18} acyloxy; chloro; phenyl-C_{1-9} alkyl; phenylthio; phenyl-C_{1-9} alkyl or phenylthio substituted on the phenyl ring by one to three substituents selected from C_{1-12} alkyl, hydroxy and R_{15}CO-O-; phenyl; phenyl substituted by one or two C_{1-12} alkyl groups, said alkyl groups having a maximum of 16 carbon atoms; nitro; a group of formula (b/2), (b/3) or (b/4)

$-\overset{O}{\overset{\|}{C}}-R_{11}$ (b/2)

$-CH_2-S-R_{12}$ (b/3)

$-CH(C_6H_5)CO-O-R_7$ (b/4); or

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a group of formula (a/4) or (a/5) as defined above;

with the provisos that:

a) with respect to the substituents R_{3a}, R_{4a} and R_{5a}, a maximum of two of said substituents is C_5 or C_6 cycloalkyl, C_{1-5} alkyl-C_5 or C_6 cycloalkyl, hydroxy, C_{1-22} alkoxy, optionally substituted phenoxy, C_{1-18}-acyloxy or chloro, and only one of said substituents may be optionally substituted phenyl, phenyl-C_{1-9} alkyl or phenylthio, nitro or a group of formula (b/2), (b/3), (b/4), (a/4) or (a/5), provided that only the R_{3a} substituent can be a group of formula (b/3) or (b/4) and only the R_{3a} or R_{5a} substituent can be a group of formula (a/4) or (a/5); and

b) when R_{11} in the group of formula (b/2) is other than hydrogen, such group is adjacent to a hydroxy group;

each R_7, independently, is hydrogen; C_{1-18} alkyl; alkyl-O-alkylene having a maximum of 18 carbon atoms; alkyl-S-alkylene having a maximum of 18 carbon atoms; di-C_{1-4}-alkylamino-C_{1-8} alkyl; C_{5-7} cycloalkyl;

phenyl; or phenyl substituted by one to three C_{1-12} alkyl groups, said alkyl groups having a maximum of 18 carbon atoms;

either each R_8, independently, is hydrogen; C_{1-18} alkyl; C_5 or C_6 cycloalkyl; C_{1-5} alkyl-C_5 or C_6 cycloalkyl; phenyl; phenyl substituted by one or two C_{1-12} alkyl groups, said alkyl groups having a maximum of 16 carbon atoms; or a group of formulae (d/1), (d/2) or (d/3)

$-CH_2CH_2OH$ (d/1)

$-CH_2CH_2OC_{1-18}alkyl$ (d/2)

$-CH_2CH_2-O-\overset{O}{\overset{\|}{C}}-R_{11}$ (d/3) ;

or both R_8's, together with the nitrogen atom, form an unsubstituted piperidine or morpholine ring;

R_9 has one of the significances of R_8;

R_{9a} is hydrogen; C_{1-18} alkyl; or a group of formula (d/1), (d/2) or (d/3) as defined above;

R_{10a} is hydrogen; C_{1-18} alkyl; C_5 or C_6 cycloalkyl; C_{1-5} alkyl-C_5 or C_6 cycloalkyl; phenyl; phenyl substituted by one or two C_{1-12} alkyl groups, said alkyl groups having a maximum of 16 carbon atoms; or benzyl;

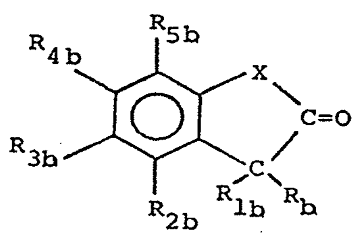
R_{11} is hydrogen; C_{1-22} alkyl; C_{5-7} cycloalkyl; phenyl; phenyl-C_{1-6} alkyl; or phenyl or phenyl-C_{1-6} alkyl substituted on the phenyl ring by one or two C_{1-12} alkyl groups, said alkyl groups having a maximum of 16 carbon atoms;

R_{12} is C_{1-18} alkyl; 2-hydroxyethyl; phenyl; or C_{1-9} alkylphenyl;

R_{15} is C_{1-22} alkyl; or phenyl;

and n is 0, 1 or 2,

said compound of formula Ia contains only two benzofuran(2)one or indolin(2)one nuclei, wherein the substituents on the two benzofuran(2)one or indolin(2)one nuclei are the same or different; and the bridged poly-benzofuran(2)one or poly-indolin(2)one compound is of formula Ib,



Ib

in which X is as defined above, and

each of R_b, R_{1b}, R_{2b}, R_{3b}, R_{4b} and R_{5b} has the significances corresponding to R_a, R_{1a}, R_{2a}, R_{3a}, R_{4a} and R_{5a}, respectively, as defined above with the provisos that:

B

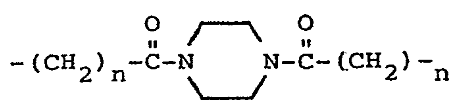
1) the molecule is free from groups of formulae (aa/1), (a/3), (a/7), (E_3) and (E_5); and

2) one of R_{1b}, R_b and R_{1b} together, R_{3b} or R_{5b} is a polyvalent bridging group linked to one or more further corresponding benzofuran(2)one or indoline(2)one nuclei,

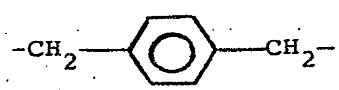
said compound of formula Ib contains two or more benzofuran(2)one or indolin(2)one nuclei, wherein the substituents on the benzofuran(2)one or indolin(2)one nuclei are the same or different.--

4. A process according to Claim 3 in which when R_{1b} is a bridging group linked to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei, it is a group of the formulae,

$$-(CH_2)_n-\overset{O}{\overset{\|}{C}}-Z-A\left(-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_{\overline{n}}\right)_w \qquad (e/1)$$

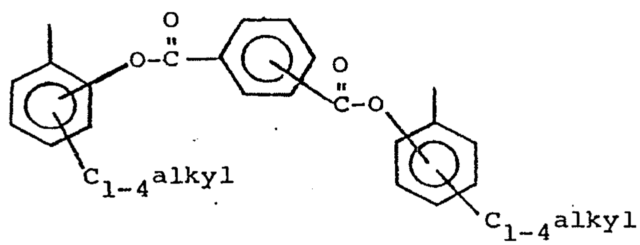


(e/4)

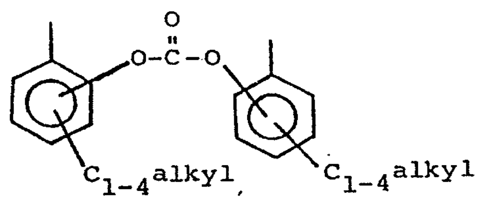


(e/5)

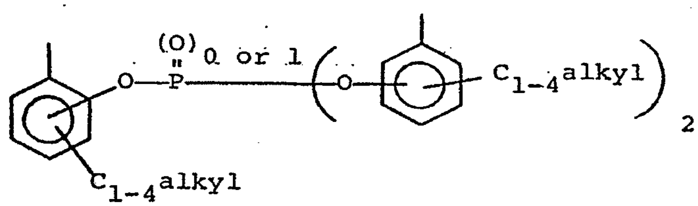
$$-C_mH_{2m}- \qquad (e/6)$$



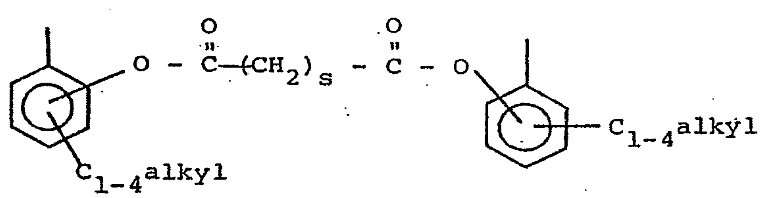
(e/7)



(e/7a)

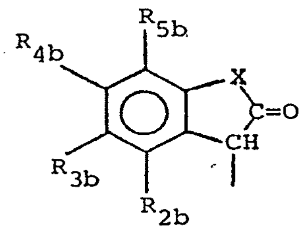


(e/7b)



(e/8)

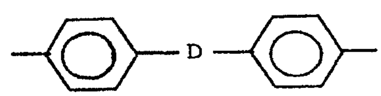
in which the free valencies are attached to one or more groups of the formula E_1



E_1

wherein each of X, R_{2b}, R_{3b}, R_{4b} and R_{5b} is as defined in Claim 28,

A is a 2 to 6 valent saturated alkylene group; a 2 to 6 valent saturated alkylene group containing one or more bridging members selected from the group consisting of a sulfur atom, an oxygen atom, a nitrogen atom and a cyclohexylene group; a 2- or 3-valent benzene group; or, when both Z's are -O-, is a group of the formula (e/16)



(e/16),

whereby when A is a 3-, 4-, 5- or 6-valent saturated alkylene group, each further free valence is bound to -OH, $-NHR_{10}$ or a group of the formula

$$-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_1$$

B.

with the proviso that any free valence on nitrogen in A itself is attached to a group of the formula

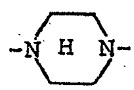
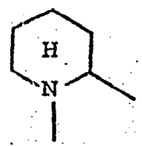
$$-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_1 \quad ;$$

w is an integer 1 to 5;

each Z, independently, is -O- or $-\overset{|}{N}-R_{10}$;

either R_{10} has one of the significances of R_{10a} as defined in Claim 28,

or R_{10}, together with the nitrogen atom, is a ring of the formula

 or  ;

D is a direct bond; -O-; -S-; $-SO_2-$; $>C{=}O$; or $-\overset{R_{13}}{\underset{R_{13}}{\overset{|}{\underset{|}{C}}}}-$

where each R_{13}, independently, is hydrogen, C_{1-16} alkyl, phenyl or a group of formula (a/4) or (a/5) as defined in Claim 28, with the proviso that when both R_{13}'s are C_{1-16} alkyl, the alkyl groups contain a maximum of 16 carbon atoms;

n is as defined in Claim

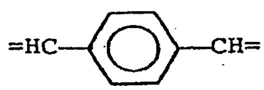
m is an integer 2 to 10; and

s is 0 or an integer 1 to 12;

when R_b and R_{1b} together is a bridging group linked to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei, it is a group of the formulae

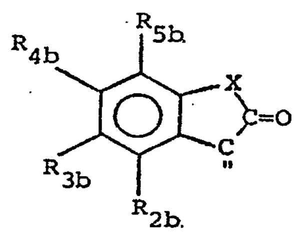
$$= \overset{R_{16}}{\overset{|}{C}} - \overset{O}{\overset{\|}{C}} - Z - A \left(- Z - \overset{O}{\overset{\|}{C}} - \overset{R_{16}}{\overset{|}{C}} = \right)_w \qquad (e/9)$$

$$=HC-(C_pH_{2p})-CH= \qquad (e/12)$$



, (e/13)

in which the free valencies are attached to one or more groups of the formula E_{1a}



E_{1a}

wherein each of X, R_{2b}, R_{3b}, R_{4b} and R_{5b} is as defined in Claim 28,

A, Z, w and R_{10} are as defined above, whereby when A is a 3-, 4-, 5- or 6-valent saturated alkylene group, each further free valence is bound to -OH, $-NHR_{10}$ or a group of the formula

$$-Z-\overset{O}{\overset{\|}{C}}R_{16}=E_{1a} \quad ,$$

with the proviso that any free valence on nitrogen in A itself is attached to a group of the formula

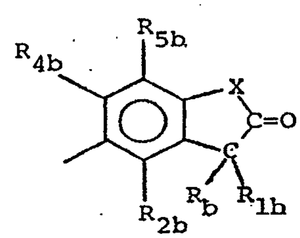
$$-\overset{O}{\overset{\|}{C}}-CR_{16}=E_{1a} \quad ;$$

p is 0 or an integer 1 to 10; and

R_{16} is hydrogen or methyl;

when R_{3b} is a bridging group linked to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei, it is -O-, -S-, $-SO_2-$, $>C=O$, $-\underset{R_{13}}{\overset{R_{13}}{C}}-$, where R_{13} is as defined above,

or a group (e/1) or (e/4) as defined above, in which the free valencies are attached to one or more groups of the formula E_{3b}



E_{3b}

wherein each of X, R_b, R_{1b}, R_{2b}, R_{4b} and R_{5b} is as defined in Claim 28,

A, Z, w and R_{10} are as defined above, whereby when A is a 3-, 4-, 5- or 6-valent

B

saturated alkylene group, each further free valence is bound to -OH, NHR_{10} or a group of the formula

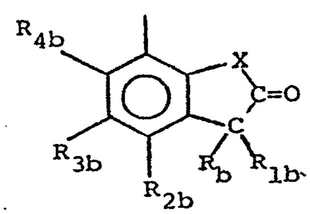
$$-Z-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_{3b} \quad ,$$

with the proviso that any free valence on nitrogen in A itself is attached to a group of the formula

$$-\overset{O}{\overset{\|}{C}}-(CH_2)_n-E_{3b} \quad ; \text{ and}$$

n is as defined in Claim 28; or

when R_{5b} is a bridging group linked to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei, it is -S-, $-\underset{R_{13}}{\overset{R_{13}}{\overset{|}{\underset{|}{C}}}}-$, where R_{13} is as defined above, or a group (e/1) or (e/4) as defined above, in which the free valencies are attached to one or more groups of the formula E_{5b}



E_{5b}

wherein each of X, R_b, R_{1b}, R_{2b}, R_{3b} and R_{4b} is as defined in Claim 3 ,

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A, Z, w and R_{10} are as defined above, whereby when A is a 3-, 4-, 5- or 6-valent saturated alkylene group, each further free valence is bound to -OH, $-NHR_{10}$ or a group of the formula

$$-Z-\overset{\displaystyle O}{\overset{\|}{C}}-(CH_2)_n-E_{5b} \quad ,$$

with the proviso that any free valence on nitrogen in A itself is attached to a group of the formula

$$-\overset{\displaystyle O}{\overset{\|}{C}}-(CH_2)_n-E_{5b} \quad ; \text{ and}$$

n is as defined in Claim 3 ;

with the proviso that only one of R_{1b}, R_b and R_{1b} together, R_{3b} or R_{5b} is a polyvalent bridging group linked to one or more further corresponding benzofuran(2)one or indolin(2)one nuclei, the substituents in the nuclei being the same or different.

5. A process according to Claim 4, in which when R_{3b} or R_{5b} is a bridging group, X is -O- in all cases.

6. A process according to Claim 3, in which X is -O-.

7. A process according to Claim 4, in which R_a is hydrogen or (aa/1),

R_{1a} is R_{1a}', where R_{1a}' is (a/7) or R_1', where R_1' is hydrogen, C_{1-18}alkyl, phenyl optionally substituted by one or two C_{1-8}alkyl groups and/or a hydroxyl group; (a/4) or (a/5).

R_{1ax} is C_{1-18}alkyl or phenyl optionally substituted by one or two C_{1-18}alkyl groups and/or a hydroxyl group, each of R_{2a} and R_{2b} is hydrogen or C_{1-4}alkyl,

R_{3a} is R_{3a}', where R_{3a}' is E_3 or R_3', where R_3' is hydrogen or C_{1-9}alkyl, each of R_{4a} and R_{4b} hydrogen or C_{1-4}alkyl,

R_{5a} is R_{5a}', where R_{5a}' is E_5 or R_5', where R_5' is hydrogen or C_{1-8}alkyl,

R_b is hydrogen or together with R_{1b} is (e/9) or (e/13)

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R_{1b} is R_{1bx} where R_{1bx} is either R_1' or is (e/1), (e/4), (e/6), (e/7), (e/7a), (e/7b) or (e/8),

R_{3b} is R_{3bx}, where R_{3bx} is hydrogen or C_{1-9}alkyl or $-S-E_{3b}$, $-\overset{(R_{13}')_2}{C}-E_b$, (e/1) or (e/4)

in which each R_{13}', independently, is hydrogen, (C_{1-4})alkyl or (a/4) in which R_7 is hydrogen or (C_{1-18})alkyl, with the proviso that when one of R_{13}' is (a/4) the other R_{13}' is other than (a/4),

R_5 is R_{5bx}, where R_{5bx} is hydrogen, C_{1-8}alkyl or $-S-E_{5b}$, $-\overset{(R_{13}')}{C}-E_{5b}$ or (e/1), and

R_{10} is hydrogen, C_{1-2}alkyl or phenyl.

8. A process according to Claim 7, in which X is -O-.

9. A process according to Claim 7, in which any R_1' is C_{1-18}alkyl or phenyl optionally substituted by one or two C_{1-8}alkyl groups and/or a hydroxyl group.

10. A process according to Claim 7, in which X is -O- and in each benzofuran(2)one nucleus is identical with the exception of the case where R_{1a} is (a/7) where R_{1a} in the second nucleus is R_{1ax}.

11. A process according to Claim 10, in which R_{1a} is (aa/1).

12. A process according to Claim 10, in which R_{1a} is (a/7).

13. A process according to Claim 7,

in which R_b is hydrogen.

14. A process according to Claim 1, in which the polymeric material is polypropylene, polyethylene, ethylene/propylene copolymers, PVC, polyesters, polyamides, polyurethanes, polyacrylonitrile, ABS terpolymers, terpolymers of acrylic ester, styrene and acrylonitrile, copolymers of styrene and acrylonitrile, styrene/butadiene copolymers, polybutylene or polystyrene.

15. A process according to Claim 1, in which the polymeric material is polypropylene.

16. A process according to Claim 1, in which the polymeric material is polyethylene or an ethylene/propylene copolymer.

17. A process according to Claim 16, in which the polymeric material is high density (HD) polyethylene.

18. A process according to Claim 1, in which from 0.01 to 5% , based on the weight of the polymeric material, of the benzofuran(2)one or indolin(2)one compound is incorporated in the polymeric material to be stabilised.

19. A process according to Claim 1, in which the benzofuran(2)one or indolin(2)one compound is incorporated into the polymeric material by melt blending.

20. A process according to Claim 1, in which 5 to 90% of the benzofuran(2)one or indolin(2)one compound is incorporated in the polymeric material to form a stabilized master batch.

21. A process according to Claim 1, comprising incorporating the benzofuran(2)one or indolin(2)one compound

together with

either (i) a stabiliser of the sterically hindered phenol type

or (ii) a sulphur-containing or phosphorus-containing stabiliser

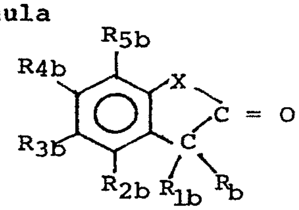
or (i) and (ii),

into the polymeric material to be stabilised.

22. Polymeric organic material whenever stabilised with a benzofuran(2)one or indolin(2)one compound as defined in Claim 1.

23. A benzofuran(2)one or indolin(2)one compound, in which the 3-, 5-, 6- or 7-position of the benzofuran(2)one or indolin(2)one nucleus is attached to the same position of 1 to 5 further such nuclei through a 2 to 6 valent bridge member.

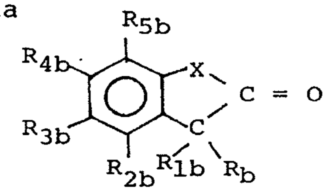
24. A benzofuran(2)one compound according to Claim 23 having the formula



Ib

wherein X, R_b, R_{1b}, R_{2b}, R_{3b}, R_{4b} and R_{5b} have the meanings given in Claim 3.

25. A benzofuran(2)one compound according to Claim 23 having the formula



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wherein X, R_b, R_{1b}, R_{2b}, R_{3b}, R_{4b} and R_{5b} have the meanings given in Claim 4.